

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
17 July 2003 (17.07.2003)

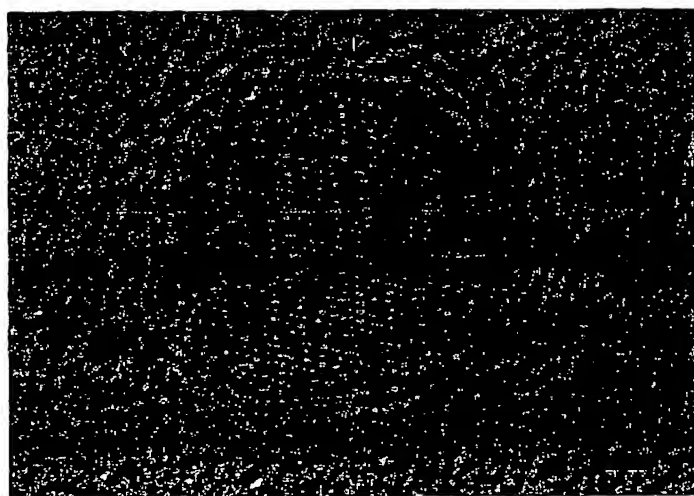
PCT

(10) International Publication Number
WO 03/057359 A2

- (51) International Patent Classification⁷: **B01J 13/02**,
A61K 9/50, C01B 31/02, C09C 3/06
- (21) International Application Number: PCT/GB03/00029
- (22) International Filing Date: 7 January 2003 (07.01.2003)
- (25) Filing Language: English
- (26) Publication Language: English
- (30) Priority Data:
0200259.0 7 January 2002 (07.01.2002) GB
- (71) Applicant (for all designated States except US): **THE UNIVERSITY OF READING** [GB/GB]; PO Box 217, Whiteknights, Reading, Berkshire RG6 6AH (GB).
- (72) Inventors; and
- (75) Inventors/Applicants (for US only): **TSANG, Shik, Chi** [GB/GB]; Department of Chemistry, University of Reading, Reading, Berkshire RG6 6AD (GB). **CHAN, Henry**,
- Bo, So [GB/GB]; 10 Longcroft Grove, Audenshaw, Manchester, Greater Manchester M34 5SD (GB).
- (74) Agents: **PAGET, Hugh, C., E. et al.**; Mewburn Ellis, York House, 23 Kingsway, London, Greater London WC2B 6HP (GB).
- (81) Designated States (national): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, OM, PH, PL, PT, RO, RU, SC, SD, SE, SG, SK, SL, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, YU, ZA, ZM, ZW.
- (84) Designated States (regional): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HU, IE, IT, LU, MC, NL, PT, SE, SI, SK, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

[Continued on next page]

(54) Title: MICROPARTICLES AND METHODS FOR THEIR PRODUCTION



(57) Abstract: Microparticles having a metal-containing core encapsulated in a graphitic shell containing hetero atoms are made by forming, in a liquid medium, colloidal particles containing a metal-oxo species of Fe, Co, Ni and Pd, colloiddally stabilized by a surfactant and containing source material of carbon and the hetero atoms. These particles are pyrolyzed in inert gas to yield the microparticles. In an alternative method, silica gel coated particles are formed by colloiddally stabilizing particles containing metal species and forming silica at the boundary of the stabilized particle.

BEST AVAILABLE COPY

WO 03/057359 A2



Published:

— without international search report and to be republished
upon receipt of that report

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

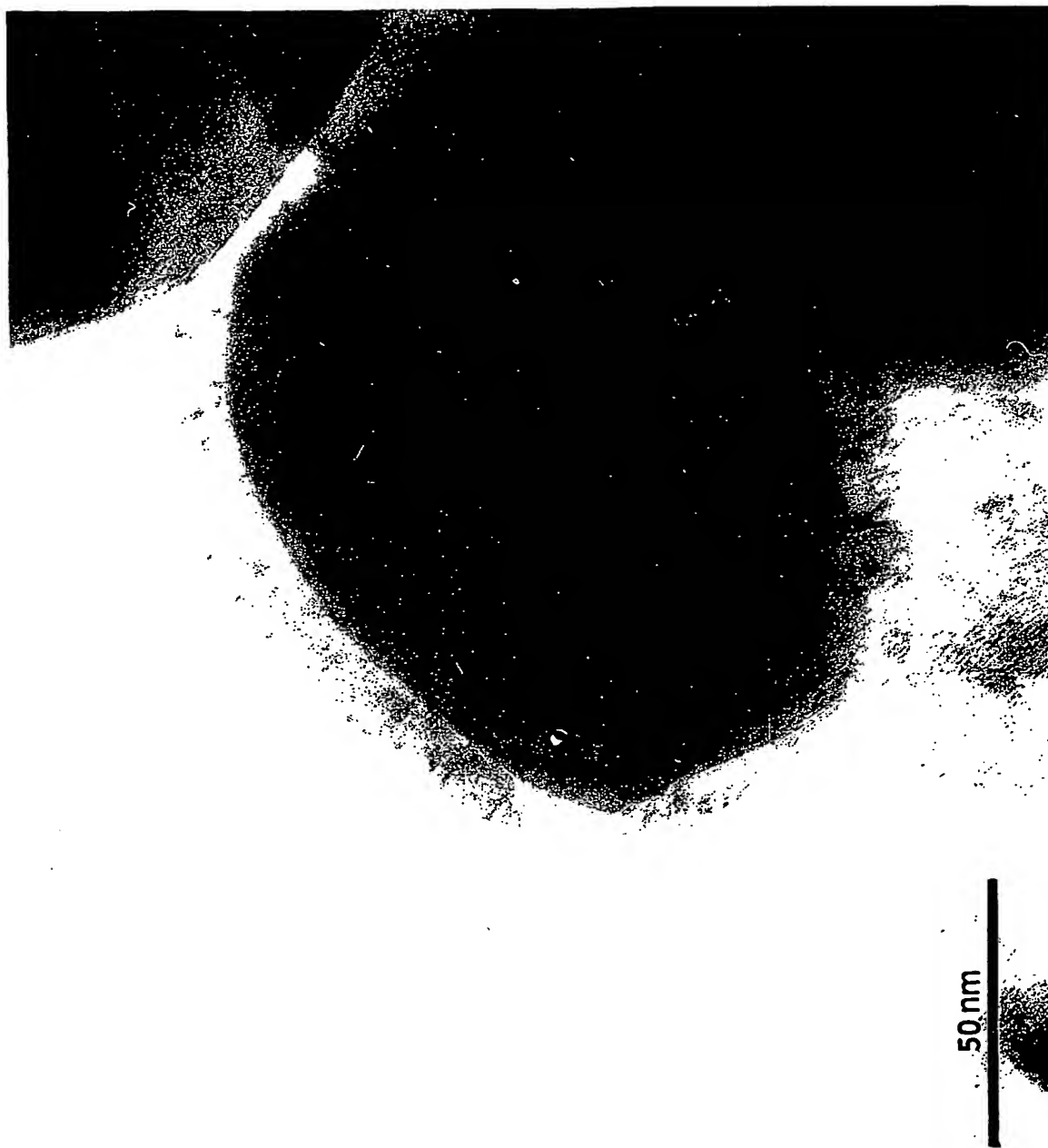
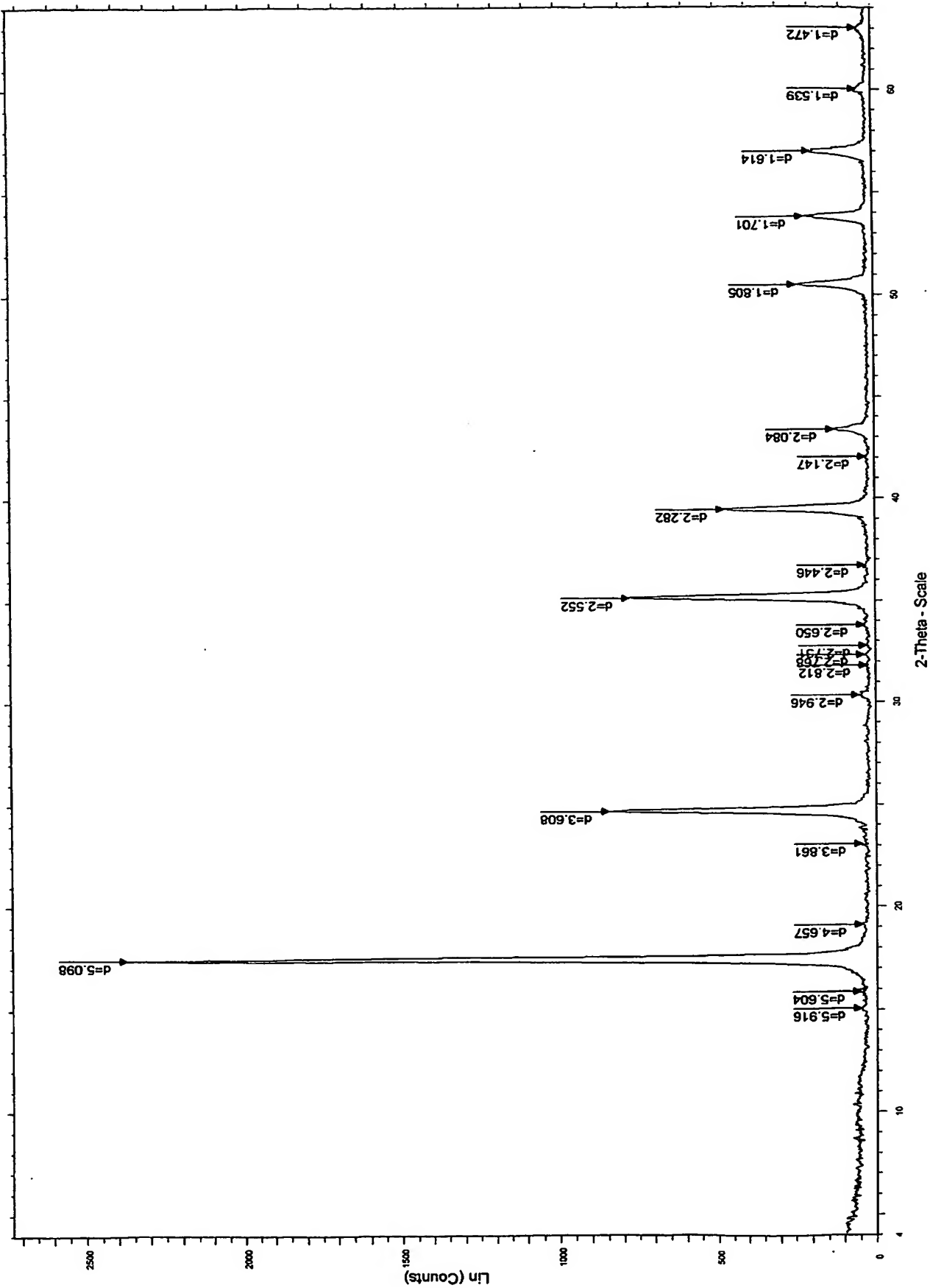


Fig. 1

Fig. 2

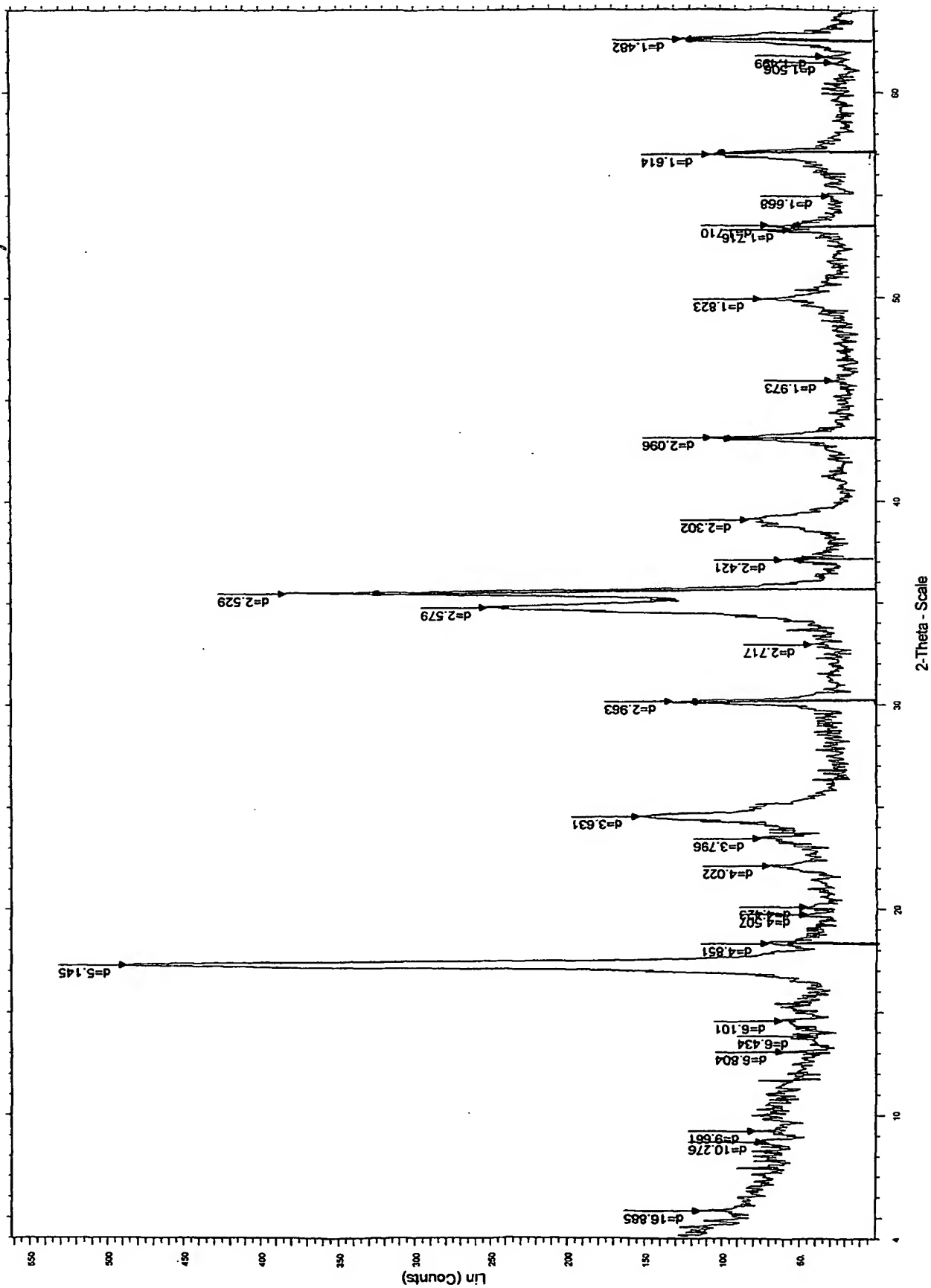
TB-HC



3/19

Fig. 3

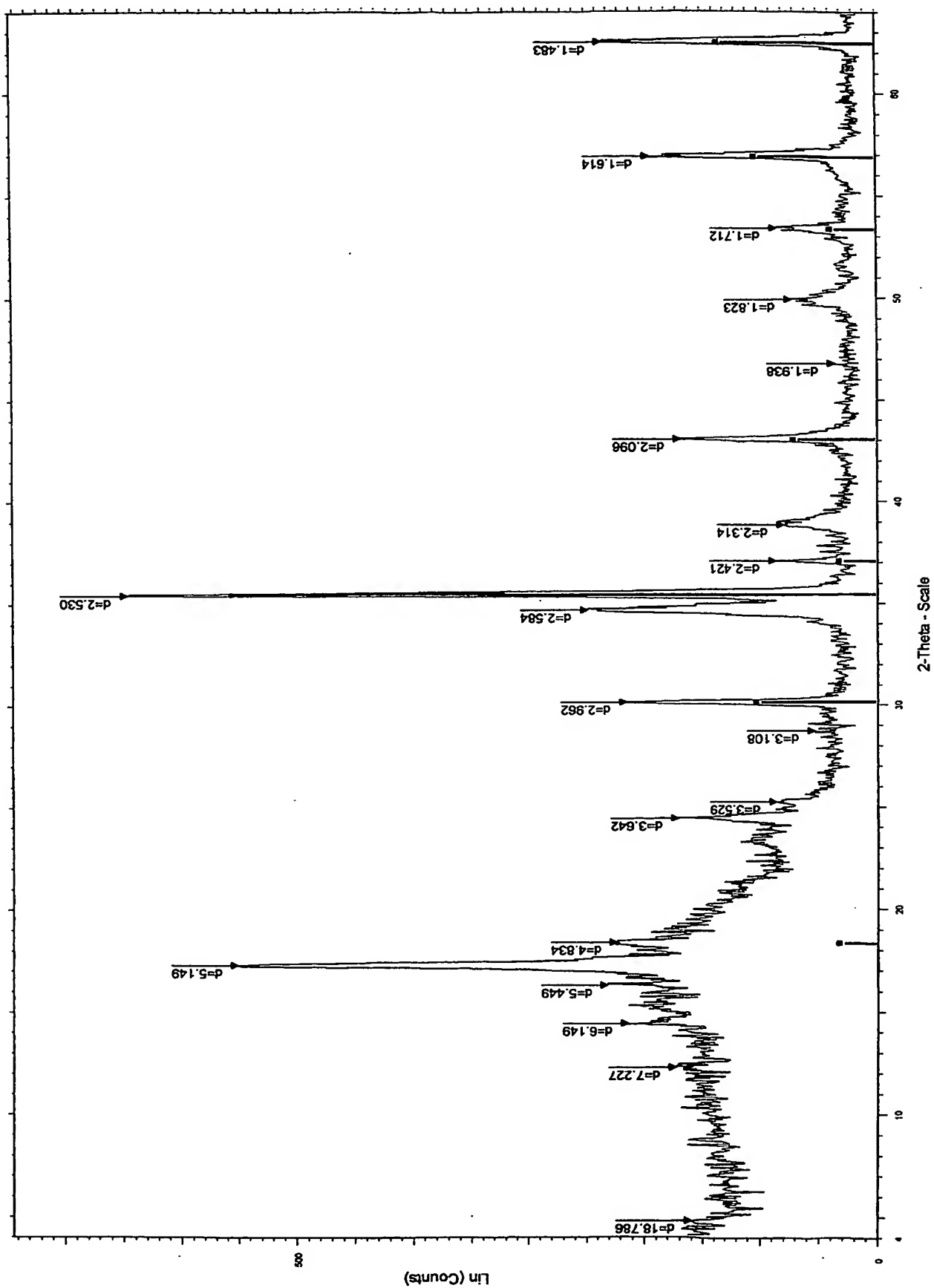
NANO-5-Fe-Product



4/19

Fig. 4

N-1-Fe/HC



5/19

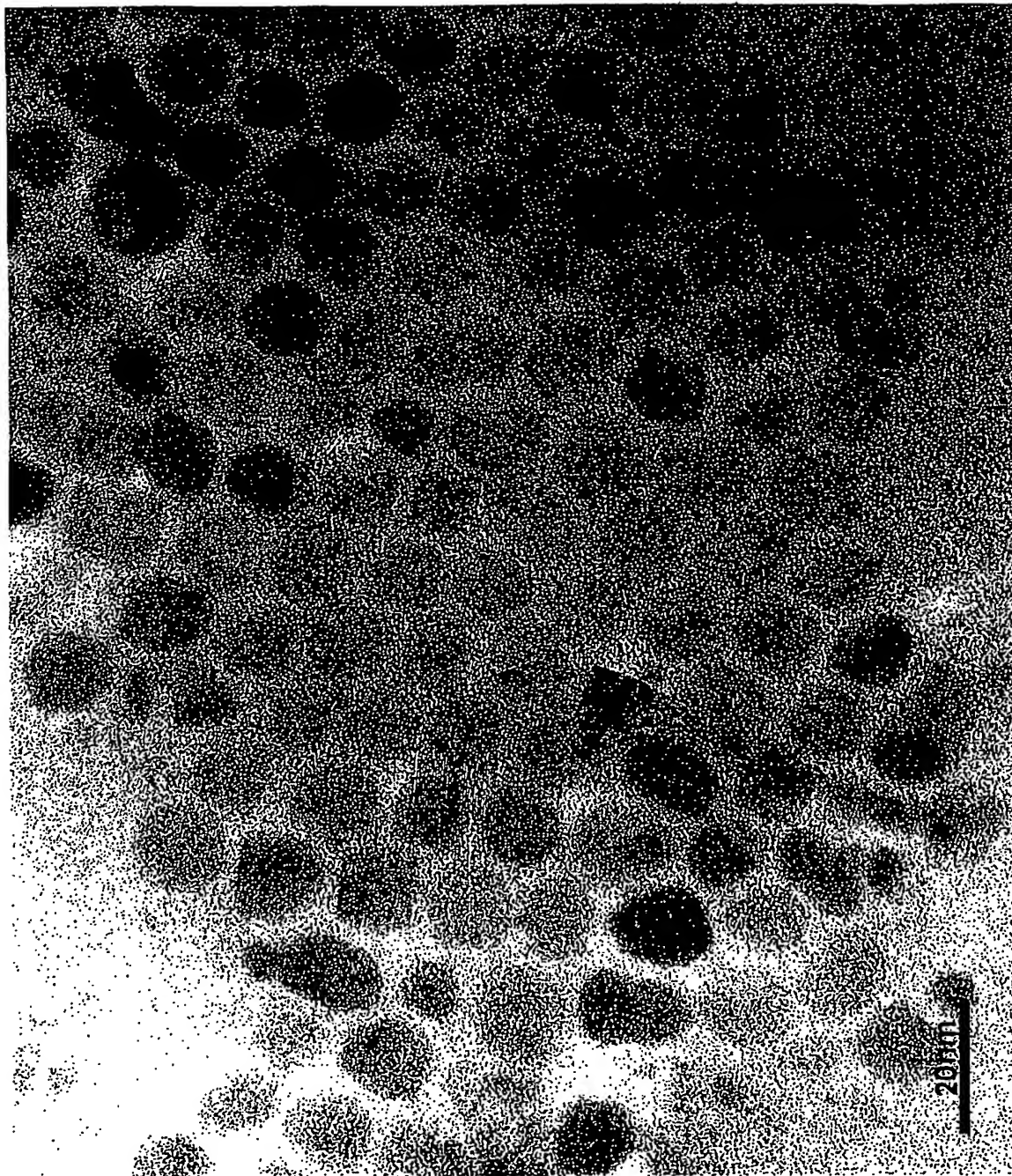


Fig. 5

6/19

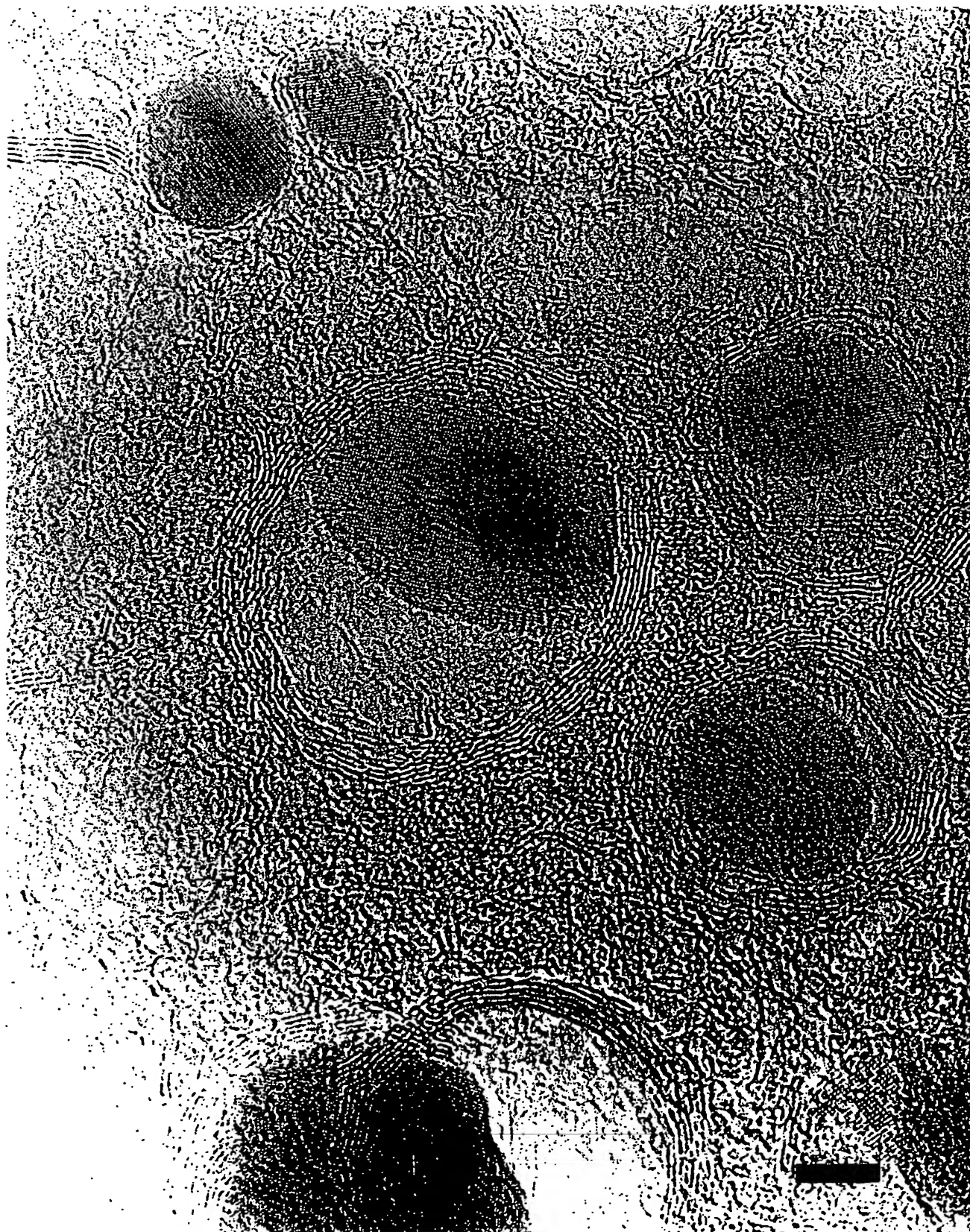


Fig. 6a

7/19

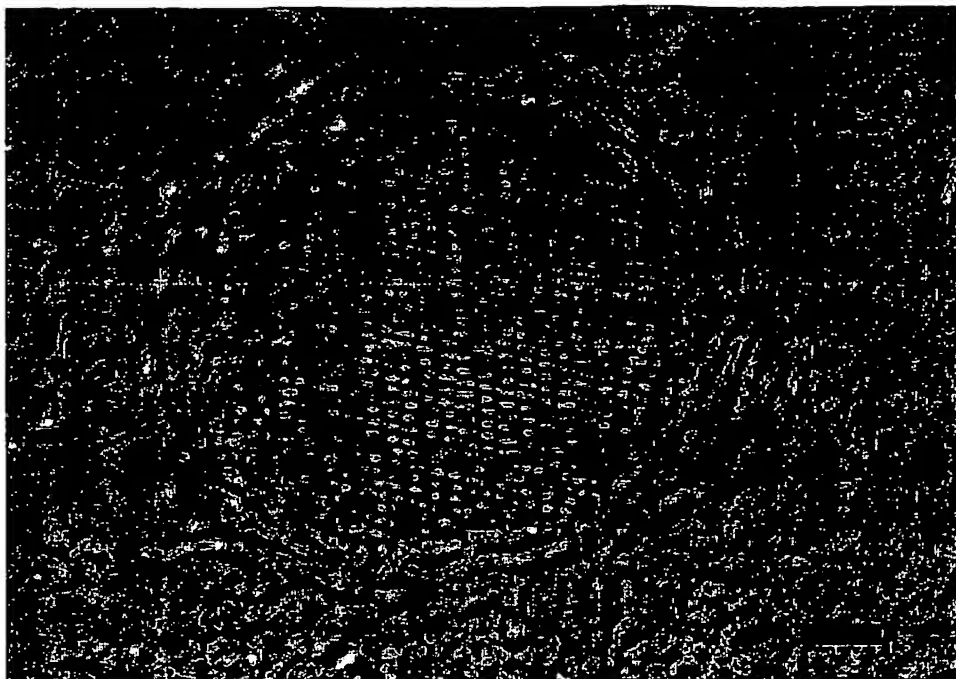


Fig. 6b

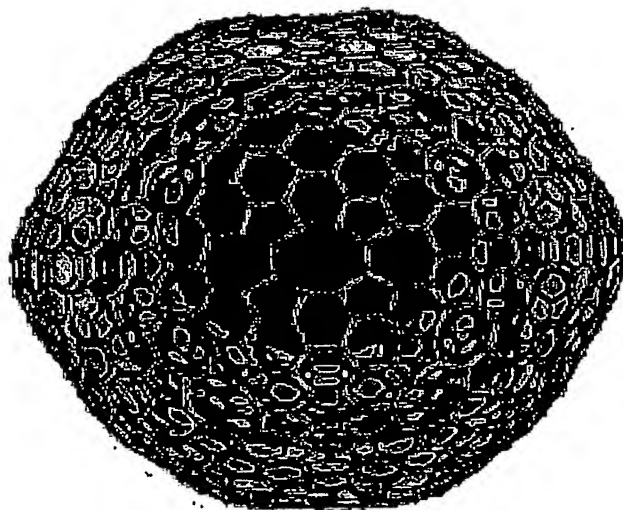


Fig. 6c

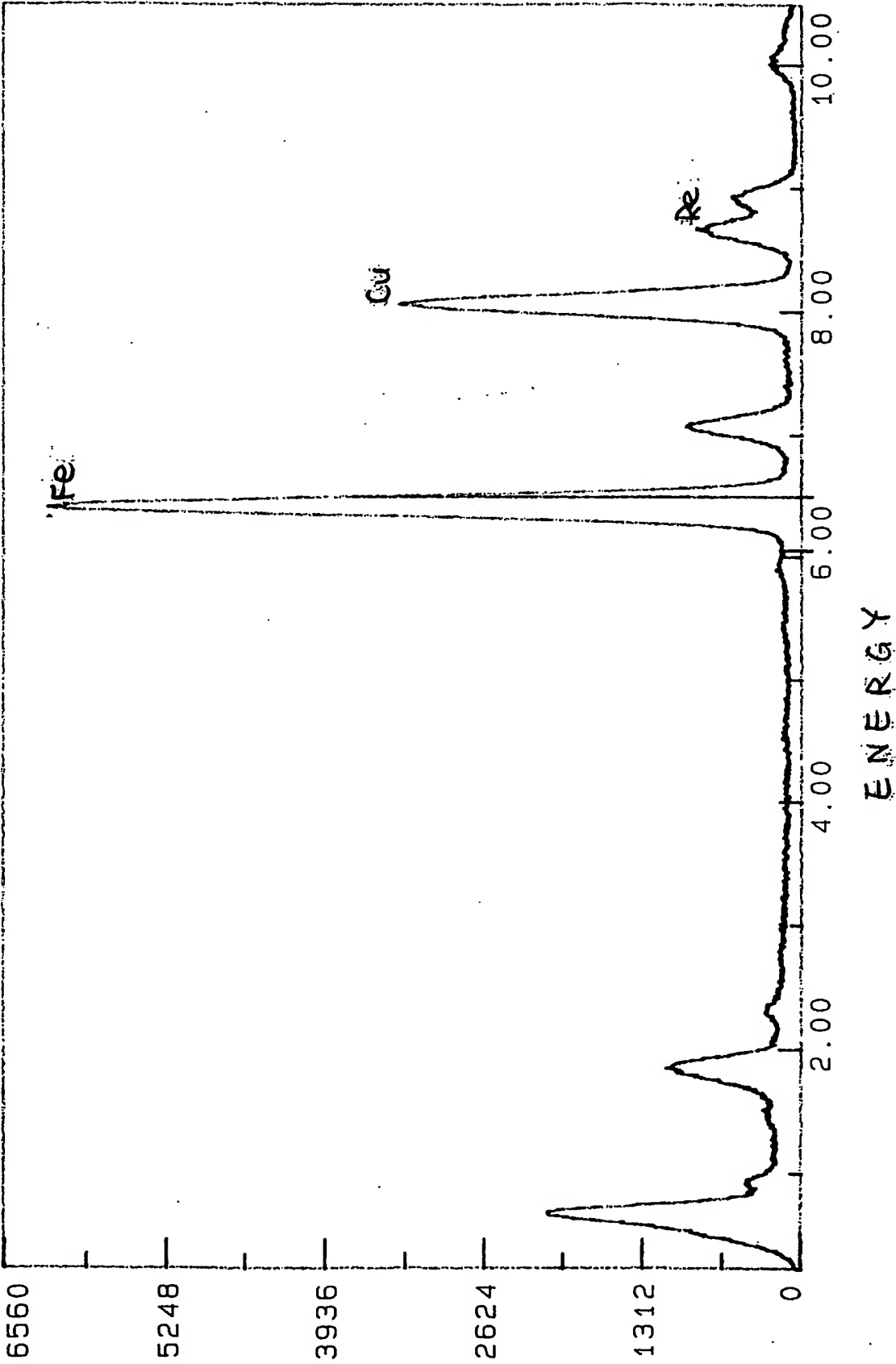


Fig. 7

9/19

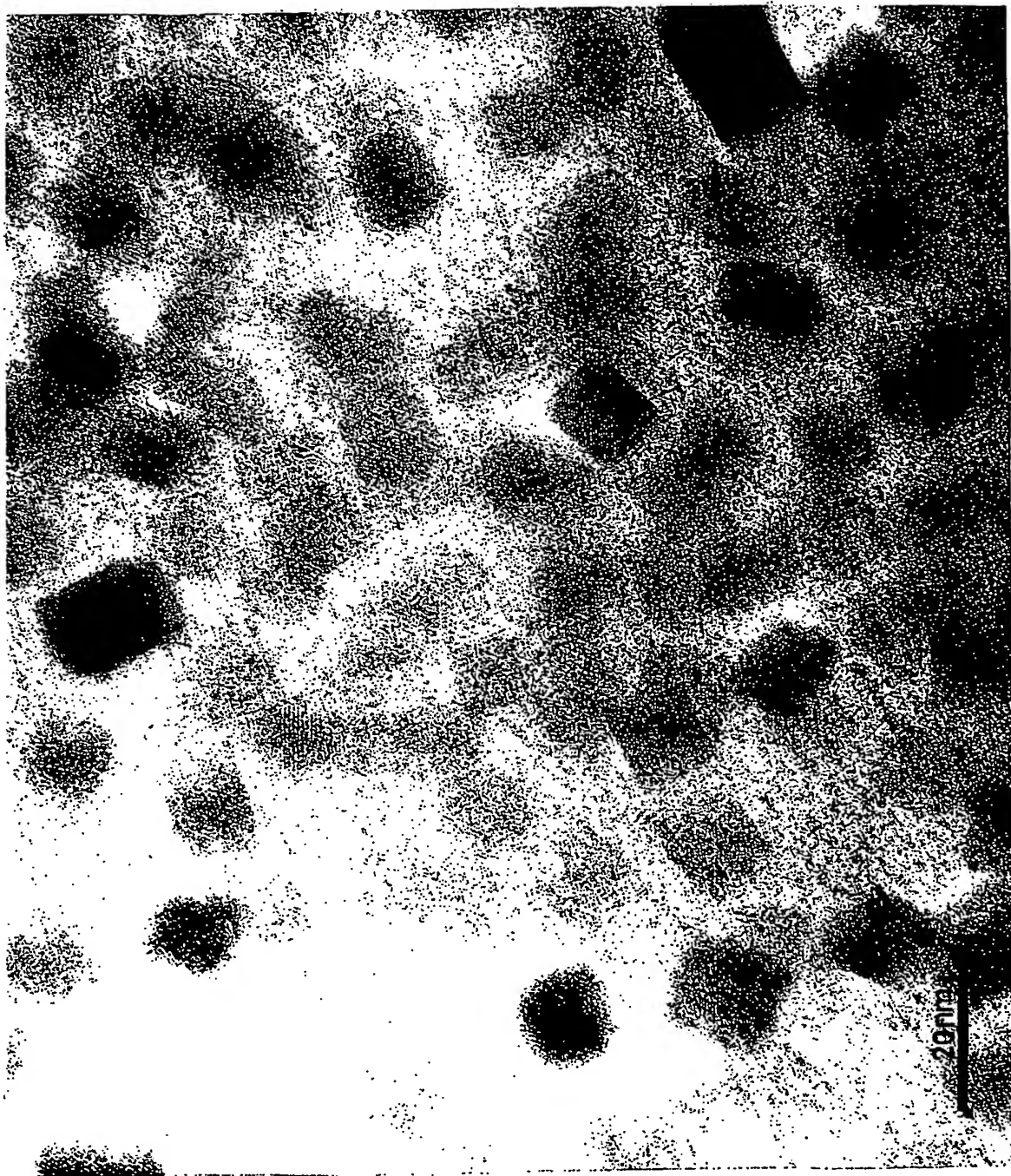


Fig. 8

10/19



Fig. 9a

11/19

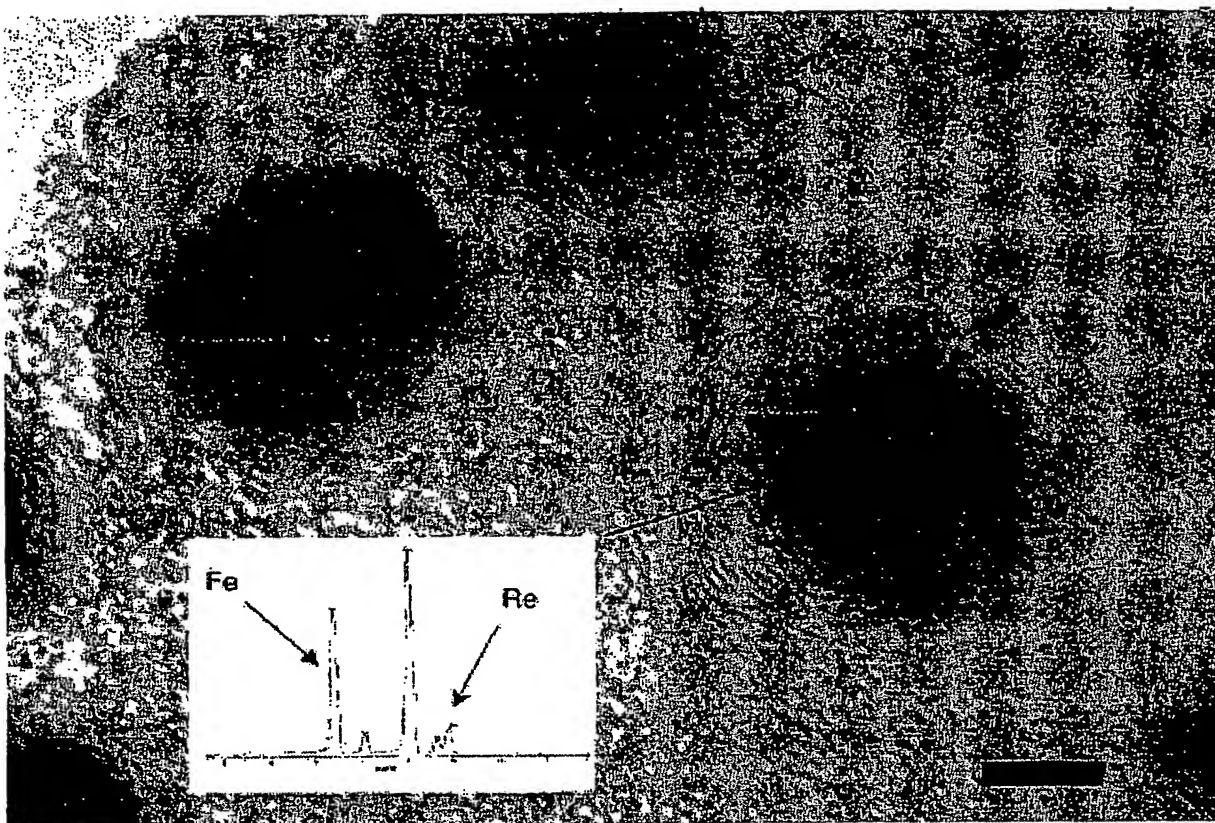


Fig. 9b

12/19

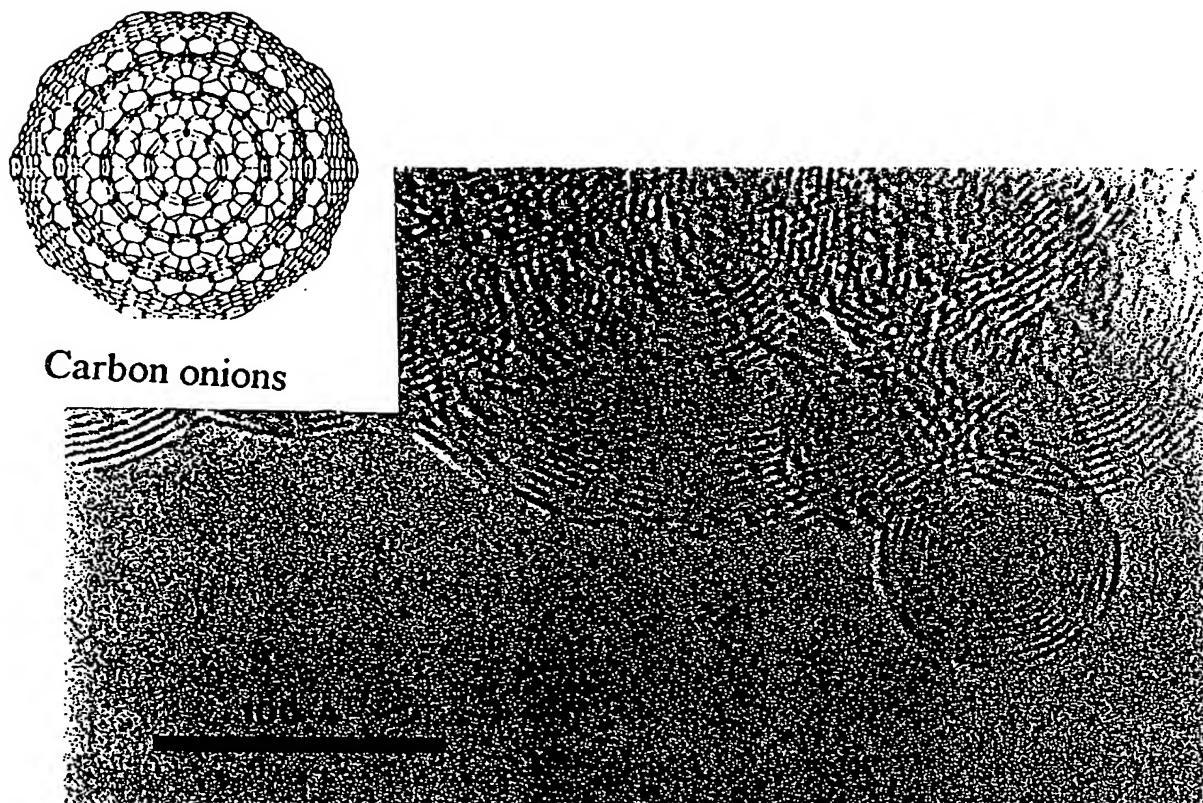


Fig. 10

13/19

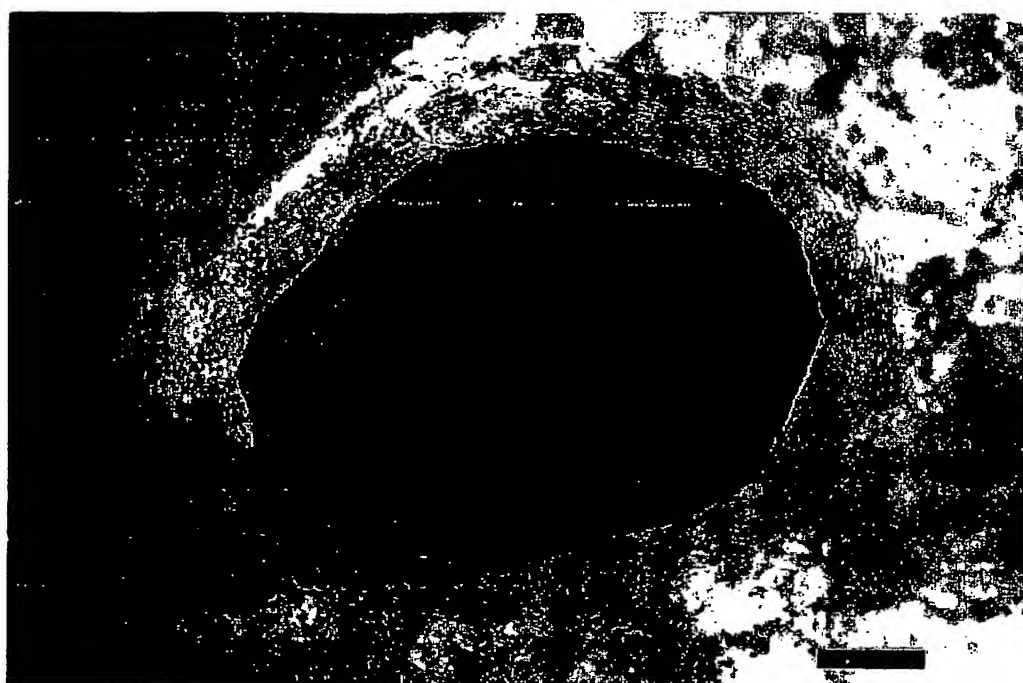


Fig. 11

14/19

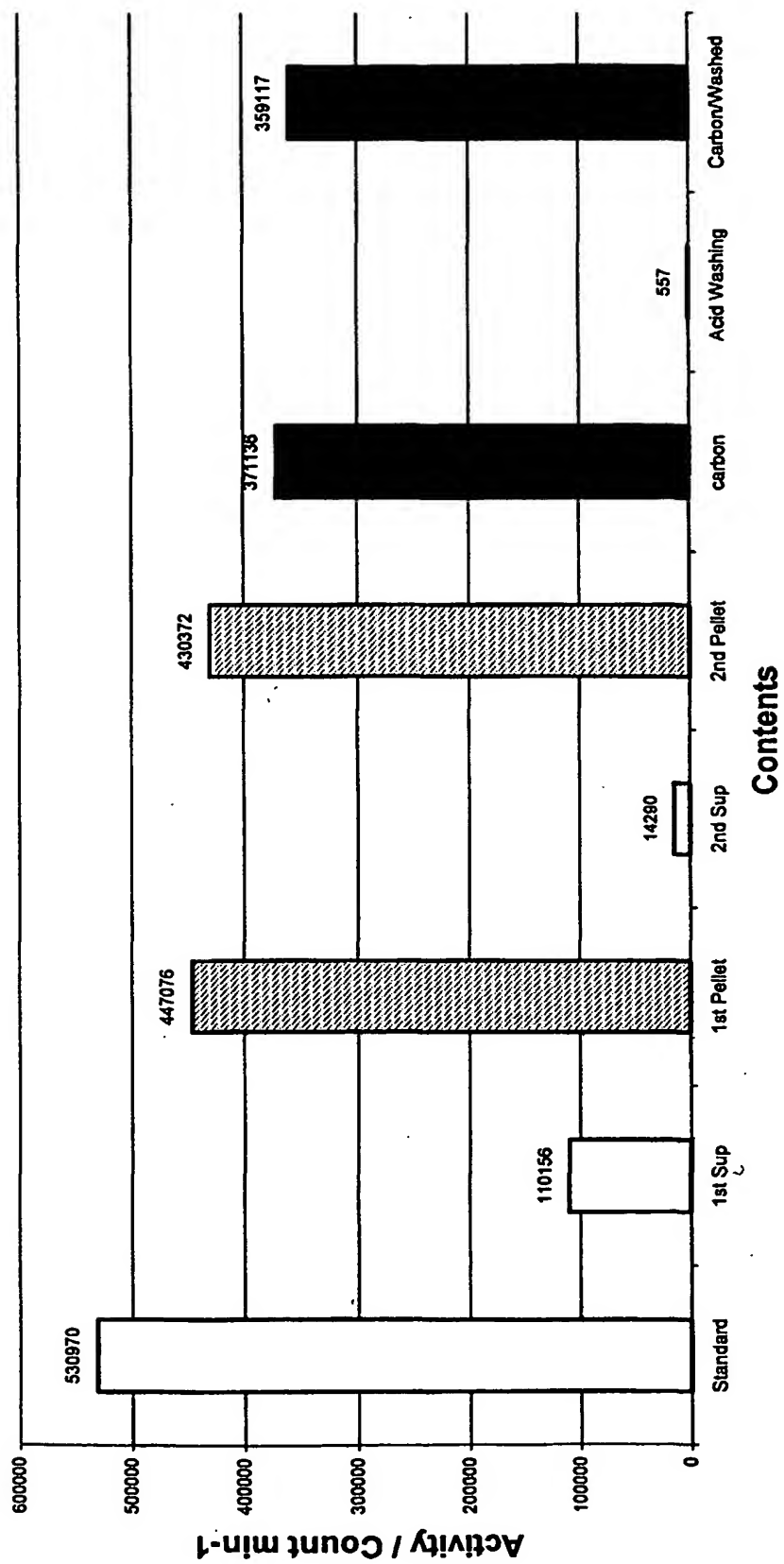


Fig. 12

15/19

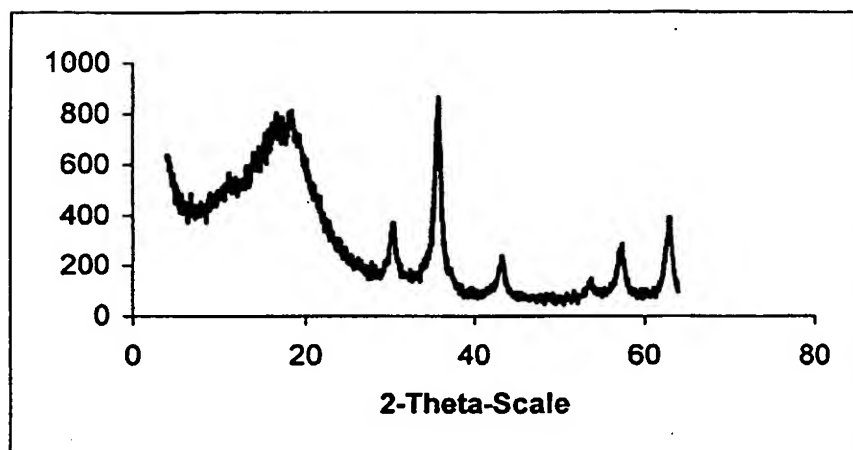


Fig. 13

16/19

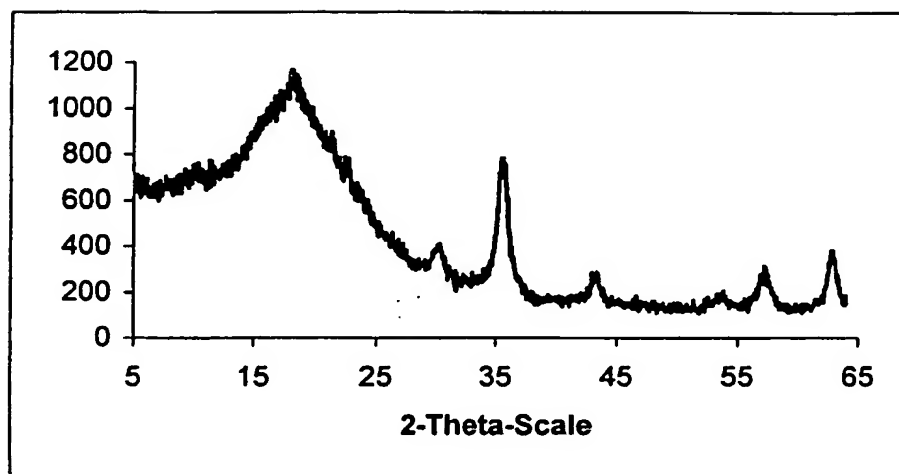


Fig. 14

17/19

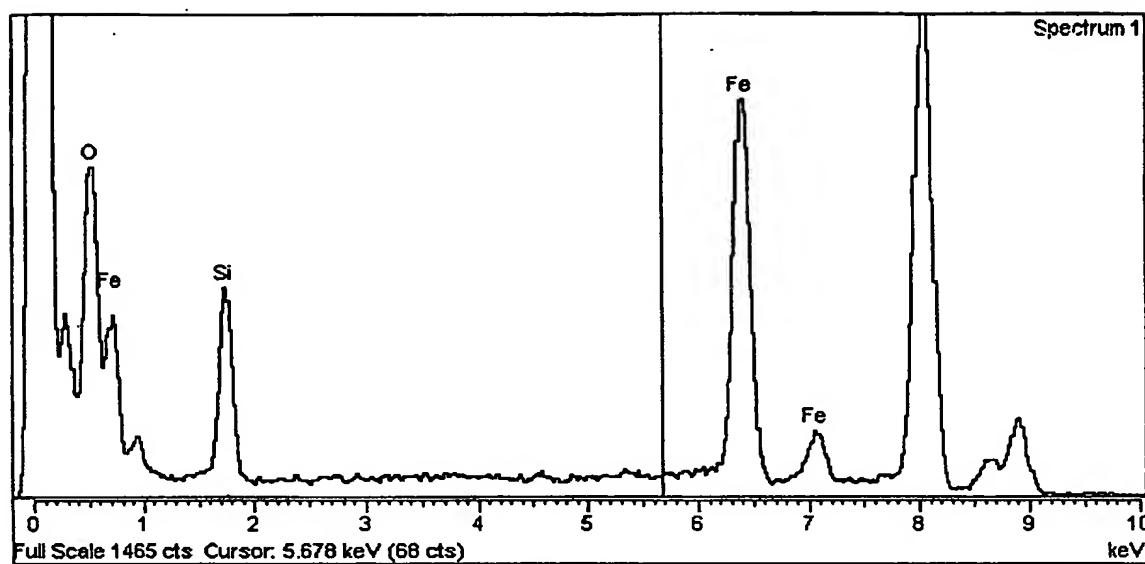


Fig. 15

18/19

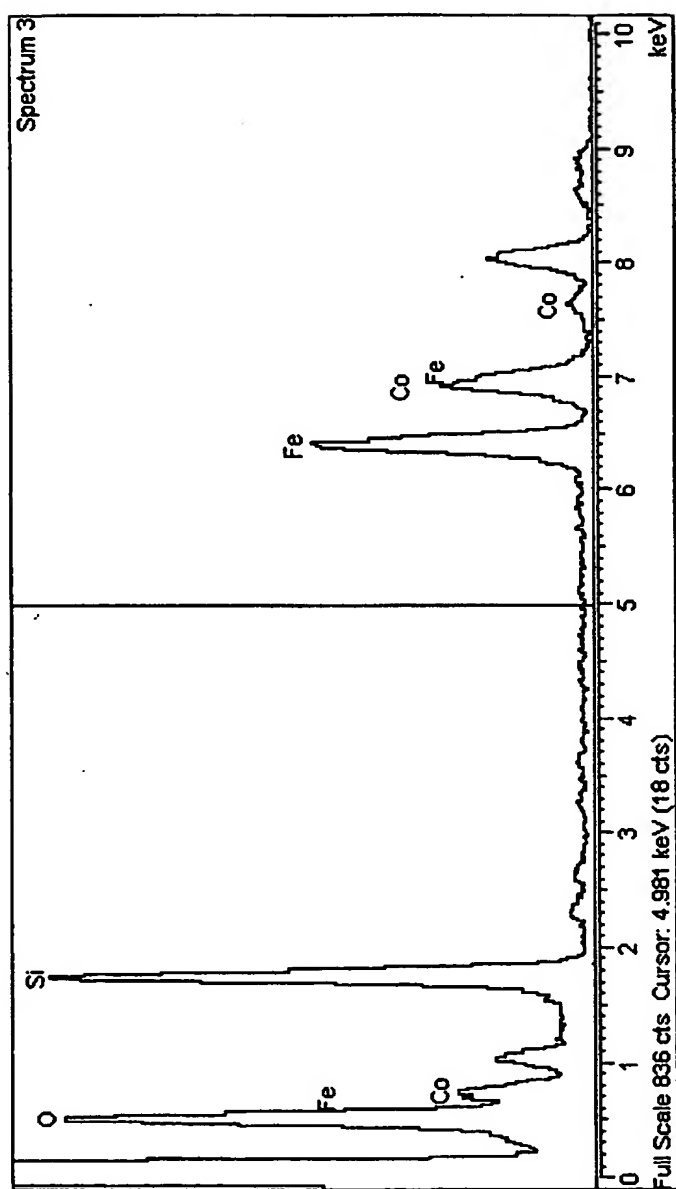


Fig. 16

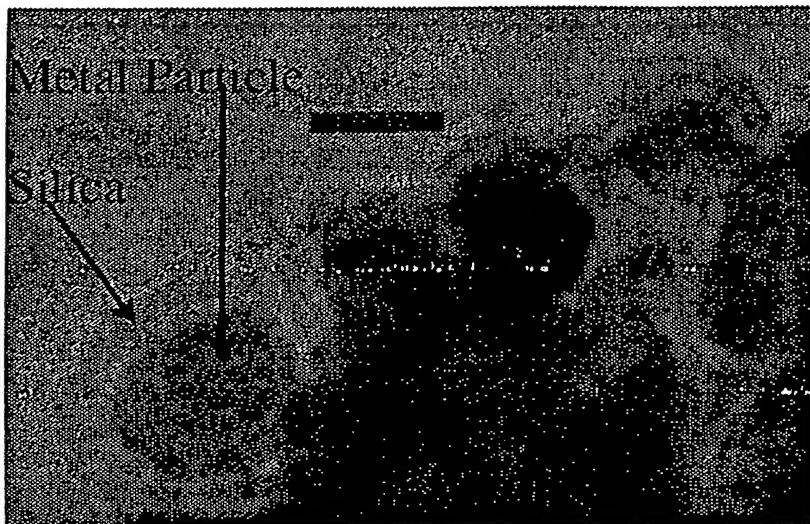


Fig. 17

Rec'd PCT/PT 7 JUN 2004

10/500842

INTERNATIONAL SEARCH REPORT

International Application No

PC., -B 03/00029

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 B01J13/02 A61K9/50 C01B31/02 C09C3/06

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 B01J A61K C01B C09C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2001/051766 A1 (GAZDZINSKI ROBERT F) 13 December 2001 (2001-12-13) paragraphs [0034]-[0037], [0284]-[0297]	12, 13
Y	paragraph [0292]	13
X	WO 02 00192 A (CARBON MEDICAL TECHNOLOGIES IN) 3 January 2002 (2002-01-03) page 2, line 27 - page 3, line 5 page 3, line 15 - line 27 page 4, line 9 - line 16 page 4, line 26 - line 30 page 9, line 25 - page 10, line 22 page 11, line 10 - line 19 page 12, line 9 - line 12 page 12, line 20 - page 13, line 27	1-15
Y	---	13

	-/--	



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

10 April 2003

Date of mailing of the international search report

23.06.03

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

Gosselin, D

INTERNATIONAL SEARCH REPORT

International Application No

PC., JB 03/00029

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 472 749 A (HWANG J-H ET AL) 5 December 1995 (1995-12-05) column 1, line 26 -column 2, line 13 ---	1-15
A	GADD G E ET AL: "The encapsulation of Ni in graphitic layers using C60 as a precursor" CARBON, XX, XX, vol. 39, no. 12, October 2001 (2001-10), pages 1769-1787, XP004299747 ISSN: 0008-6223 the whole document ---	1-15
Y	US 6 009 457 A (MOLLER MATTHEW DONALDSON) 28 December 1999 (1999-12-28) column 5, line 7-14 column 6, line 14 - line 26 column 6, line 50 -column 8, line 6 column 9, line 30 -column 10, line 18 column 11, line 40 - line 52 column 15, line 15 -column 44, line 43 -----	13

INTERNATIONAL SEARCH REPORT

International application No.
PCT/GB 03/00029

Box I Observations where certain claims were found unsearchable (Continuation of Item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of Item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

see additional sheet

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

1-15

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. Claims: 1-15

The method for encapsulating metal species in a graphitic shell according to claims 1 to 11 and microcapsules with a graphitic shell according to claims 12 to 15

2. Claims: 16-28

The method for encapsulating metal-species in a silica shell according to claims 16 to 22 and microcapsules with a silica shell according to claims 23 to 28.

INTERNATIONAL SEARCH REPORT

International Application No

PC., JB 03/00029

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 2001051766 A1	13-12-2001	US 2002103417 A1	01-08-2002
WO 0200192 A	03-01-2002	US 6355275 B1	12-03-2002
		EP 1305004 A1	02-05-2003
		WO 0200192 A2	03-01-2002
US 5472749 A	05-12-1995	AU 4282796 A	23-05-1996
		WO 9613339 A1	09-05-1996
		US 5618475 A	08-04-1997
		US 5665277 A	09-09-1997
US 6009457 A	28-12-1999	AU 730214 B2	01-03-2001
		AU 2298597 A	22-10-1997
		CA 2250855 A1	09-10-1997
		EP 0891665 A1	20-01-1999
		WO 9737476 A1	09-10-1997
		JP 2000507715 T	20-06-2000

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

☐ **BLACK BORDERS**

☐ **IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**

☒ **FADED TEXT OR DRAWING**

☐ **BLURRED OR ILLEGIBLE TEXT OR DRAWING**

☐ **SKEWED/SLANTED IMAGES**

☐ **COLOR OR BLACK AND WHITE PHOTOGRAPHS**

☐ **GRAY SCALE DOCUMENTS**

☐ **LINES OR MARKS ON ORIGINAL DOCUMENT**

☒ **REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**

☐ **OTHER:** _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.